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LOW SOLUBILITY HYDRATED CALCIUM SILICATE

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LOW SOLUBILITY HYDRATED CALCIUM SILICATE

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OWNERS: **JOHNS-MANVILLE CORPORATION**  
ISSUED: **July 5 , 1960**

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CLAIMS: [Show all claims](#)

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- 2 -

601158

This invention relates to a new hydrated calcium silicate, products thereof and methods of preparing the same.

United States Patent No. 1,574,363, issued February 23, 1926 to Calvert teaches the preparation of hydrated calcium silicates by reacting lime and a siliceous material, such as diatomaceous earth, and a water media. This patent describes a batch reaction at preferred temperatures of 90 to 105°C. to produce a filtration medium. The calcium silicate produced by such reaction conditions is identified in the art as calcium silicate hydrate I, a compound of variable composition having a  $\text{CaO/SiO}_2$  molar ratio of 0.8 to 1.5 of lime to 1 of silica and described in detail by Taylor, Journal of the Chemical Society, 163 (1950).

It is also known in the art that the product produced by a reaction such as described by Calvert, once it has been dried and dispersed, is a finely divided, absorbent powder. Accordingly, such a product may be useful as an inert carrier for organic insecticides due to its sorptive capacity, thus providing a means for producing a high concentrate flowable powder. However, there are numerous applications in the preparation of a wettable powder insecticide concentration where a product, such as produced by Calvert, is not suitable. Notably, one of these is in the preparation of a 75% wettable DDT powder which must meet the stringent specifications set up by the International Cooperation Administration.

We have now found a new hydrous silicate, and products thereof which may be prepared from its components - lime, silica and water - and which possesses novel and unique chemical and physical properties and characteristics of substantial utility. Accordingly, the present invention consists

- 3 -

601158

of a new and useful, definite hydrated calcium silicate composition, products thereof, a method of preparing the same and the use thereof.

It is one object of this invention to provide a new and definite hydrated calcium silicate and method of preparing the same.

A further object of this invention is to provide a new hydrated calcium silicate compound or product of new and unique characteristic properties, that is, a compound or product of low bulk density, greatly improved chemical stability, high liquid adsorptive capacity, and particularly a very low solubility substantially lower than that of Galvert's product, among other advantageous properties.

A further object of this invention is to provide a calcium silicate compound or product which, due to its unique properties, is particularly adaptable for use as a carrier in the preparation of a wetttable powder insecticide dispersion.

These and other objects and advantages will become apparent and more fully understood from a consideration of the following details descriptive of the invention.

The preparation of the new hydrated calcium silicate may be effected with lime and a reactive siliceous material, such as diatomaceous earth, quartz, silica gel, etc., as starting materials. Preferably finely divided starting materials, such as finely divided hydrated lime and finely divided diatomaceous silica, are employed to reduce the reaction period.

The new hydrated calcium silicate of this invention is prepared by hydrothermally reacting the starting ingredients or components in a molar ratio of about 0.67 CaO to 1 SiO<sub>2</sub>.

- 3 -

601158

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- 4 -

601158

Preferably the starting materials are initially suspended or dispersed in at least sufficient water to form a flowable slurry, e.g., about 4 to 17 parts by weight of water per part by weight of solids. It is also preferred that the slurry, suspension or the like, in which the reaction takes place has a water to solids ratio of about 8 to 50 parts by weight of water per part by weight of solids. The lime and reactive siliceous material may be dispersed individually to form separate slurries, or together to form a single slurry as is desired or appropriate.

The slurry or suspension of lime and reactive silica, prepared in accordance with the foregoing, is then hydrothermally reacted by heating said components in a reaction vessel to, and maintaining the same at a temperature of at least 182°C. (360°F.), preferably approximately 230°C. (450°F.) to effect the desired reaction. Preferably the materials in the reaction vessel are agitated so as to obtain a reasonable reaction rate, since the reaction time or duration required to obtain the desired product is dependant upon the reaction temperature and the degree of agitation. In other words, the rate of reaction is increased as the degree of agitation and the temperature are increased. Using average agitation and a minimum reaction temperature of about 182°C. a reaction time in excess of 4 hours must be used in order to obtain the desired product. At a reaction temperature of 230°C. the reaction time must be in excess of 0.25 hours and a 1 to 2 hour reaction period is preferred; however, longer reaction periods do not have a detrimental effect. In any case, a reaction temperature of at least 182°C. is considered essential. Finally, the solids produced in accordance with the foregoing are filtered from the slurry and dried.

- 4 -

601158

Preferably the starting materials are initially suspended or dispersed in at least sufficient water to form a flowable slurry, e.g., about 4 to 17 parts by weight of water per part by weight of solids. It is also preferred that the slurry, suspension or the like, in which the reaction takes place has a water to solids ratio of about 8 to 50 parts by weight of water per part by weight of solids. The lime and reactive siliceous material may be dispersed individually to form separate slurries, or together to form a single slurry as is desired or appropriate.

The slurry or suspension of lime and reactive silica, prepared in accordance with the foregoing, is then hydrothermally reacted by heating said components in a reaction vessel to, and maintaining the same at a temperature of at least 182°C. (360°F.), preferably approximately 230°C. (450°F.) to effect the desired reaction. Preferably the materials in the reaction vessel are agitated so as to obtain a reasonable reaction rate, since the reaction time or duration required to obtain the desired product is dependent upon the reaction temperature and the degree of agitation. In other words, the rate of reaction is increased as the degree of agitation and the temperature are increased. Using average agitation and a minimum reaction temperature of about 182°C. a reaction time in excess of 4 hours must be used in order to obtain the desired product. At a reaction temperature of 230°C. the reaction time must be in excess of 0.25 hours and a 1 to 2 hour reaction period is preferred; however, longer reaction periods do not have a detrimental effect. In any case, a reaction temperature of at least 182°C. is considered essential. Finally, the solids produced in accordance with the foregoing are filtered from the slurry and dried.

- 5 -

601158

The following example illustrates the preparation of our new hydrated calcium silicate, and it will be obvious that modifications can be made without departing from the spirit of the invention.

Example I

Pure silicic acid and calcium carbonate were separately ignited to 1000°C. (1832°F.). The resultant products were calcium oxide of 99.5% purity and silicon dioxide of 99.8% purity. These were mixed using 1.80 grams of silicon dioxide and 1.12 grams of calcium oxide. The mixture was placed in 14 cc of distilled water and again mixed. The resulting slurry was placed in a watertight steel container which was sealed and the container was heated to a temperature of 332°C. (630°F.) and maintained at this temperature level for four hours. Upon completion of the heating step, the container was cooled and the reacted slurry was removed, filtered and dried. Examination by X-ray analysis identified the newly formed dried material as a new and definite calcium silicate compound.

The new hydrated calcium silicate thus formed is a definite chemical compound having the formula  $2\text{CaO} \cdot 3\text{SiO}_2$  with about 1 to 2.5 mols of water of hydration, and it averages about 2 mols of water, i.e.,  $2\text{CaO} \cdot 3\text{SiO}_2 \cdot 2\text{H}_2\text{O}$ . By conforming to the foregoing procedure, especially the necessary molar ratio of lime and silica and a sufficiently high reaction temperature, since these are the major controlling factors, a substantially pure compound may be produced. The chemical formulation disclosed and claimed herein is not to be limited by any theory, but it is merely for the purpose of illustration and explanation.



- 6 -

601158

The physical and chemical properties and characteristics of the new calcium silicate composition, referred to herein after as CS-55, are substantially different from the calcium silicate hydrate I of the Calvert patent, the minerals gyrolite and truscottite or any other similar calcium silicate compound known to us. The following data obtained by standard procedures clearly illustrate and point out the distinguishing and characteristic physical and chemical properties of CS-55.

Upon X-ray analysis CS-55 manifests the following unique and characteristic patterns as compared to calcium silicate hydrate I.

Angle, 2 $\theta$	d in Å	CSHI Intensity	CS-55 Intensity
3.7	23.86	-	M
3.84	22.99	-	-
7.9	11.18	8	M
8.2	10.77	-	M
9.4	9.40	-	W
10.6	8.34	-	M
11.2	7.89	-	W
16.1	5.50	8	-
17.4	5.09	-	-
19.0	4.67	-	N
19.4	4.57	-	-
19.9	4.46	-	-
20.3	4.37	-	-
21.2	4.12	-	VB
21.5	4.13	-	-
22.0	4.04	-	W

- 7 -

601158

	Angle $2\theta$	d in $\text{\AA}$	CSHI Intensity	CS-55 Intensity
2	22.5	3.95	-	-
3	24.0	3.70	-	-
4	24.4	3.65	-	W
5	25.5	3.49	W	W
6	27.1	3.29	W	-
7	28.04	3.18	-	-
8	28.6	3.12	-	VS
9	29.0	3.08	VS	-
10	29.4	3.04	-	VS
11	29.9	2.99	-	-
12	30.0	2.98	VS	-
13	30.9	2.89	-	-
14	31.8	2.81	W	W
15	32.14	2.78	-	W
16	33.16	2.70	-	-
17	34.0	2.64	-	S
18	35.7	2.51	W	W
19	35.82	2.51	-	-
20	36.08	2.49	-	VW
21	36.88	2.43	-	-
22	37.0	2.43	-	M
23	37.3	2.41	W	-
24	39.5	2.28	M	M
25	40.4	2.23	-	M
26	VS -- Very Strong; S -- Strong; M -- Medium; W -- Weak; VW --			
27	Very Weak.			
28				
29				
30				
31				
32				
33				

- 8 -

601158

The differential thermal analysis patterns for CS-55 and calcium silicate hydrate I are illustrated in Fig. 1 of the drawings. The patterns clearly illustrate the differences in approach to, and the breaking points of the two compositions and their subsequent formation of two different compositions.

The dehydration curves for CS-55, calcium silicate hydrate I and gyrolite are shown in Fig. 2 of the drawings. The dehydration of gyrolite results in a substantially uniform and gradual loss of water before leveling off as indicated by the curve. The sorbed water loss of calcium silicate hydrate I is sudden and complete at low temperatures whereupon it becomes substantially stable at temperatures up to about 400°C. before it suddenly gives up substantially all of its water of hydration, as illustrated by the sharp rises and plateaus in its dehydration curve. Like calcium silicate hydrate I, the sorbed water loss is sudden and complete at low temperatures, i.e., about 100°C., in the dehydration of CS-55. However, unlike calcium silicate hydrate I, upon loss of its sorbed water CS-55 is substantially stable in that it gives up very little water of hydration at temperatures ranging between 100 - 1000°C. Thus, the relative stability of CS-55 in comparison with either calcium silicate hydrate I or gyrolite is clearly significant.

The carbonation rate of CS-55 is compared with that of calcium silicate hydrate I in the graph illustrated in Fig. 3 of the drawings. The noticeable difference in the carbonation rate of the two types of products is clearly illustrated by the graph.

- 9 -

601158

The solubility of the new compound or product, CS-55, is only 39 parts per million, as compared to a solubility of 101 parts per million for calcium silicate hydrate I. This particular characteristic is exceptionally noteworthy since it renders the product especially adaptable to fulfill certain requirements necessary for particular applications set forth hereinafter.

Frequently, and in many applications it is not necessary to employ the new hydrous calcium silicate CS-55 in its purest form obtainable. Therefore, it is also within the scope of this invention to prepare and utilize a product comprising an adulterated form of the compound, or a mixture of the new compound with hydrated silica and/or possibly other calcium silicate products obtained when lime and a siliceous material are employed as starting ingredients in a  $\text{CaO}$  to  $\text{SiO}_2$  molar ratio within the range of 0.05 to 0.7  $\text{CaO}$  to 1.0  $\text{SiO}_2$  and reacted together as specified above. In other words, since the advantageous and unique physical and chemical properties and characteristics of the product comprising an adulterated composition, or mixture of the new compound and hydrous silica, etc., within the foregoing  $\text{CaO}$  to  $\text{SiO}_2$  molar ratio are substantially similar to those of the pure compound and are satisfactory for many applications, it is frequently desirable and advantageous, particularly for reasons of economy, to dilute or mix the pure compound with hydrous silica by including greater quantities of a siliceous material in the initial slurry than is necessary to obtain pure  $2\text{CaO} \cdot 3\text{SiO}_2 \cdot 1$  to  $2.5 \text{H}_2\text{O}$ .

- 10 -

601158

Accordingly, it has been found that a satisfactory product may be prepared by employing a CaO to SiO<sub>2</sub> molar ratio within the range of 0.05 to 0.7 CaO to 1 SiO<sub>2</sub>, preferably, for commercial purposes, a molar ratio between 0.4 and 0.6 CaO to 1 SiO<sub>2</sub>. However, in any case the reaction temperature should be at least 182°C., (360°F.) and preferably about 232°C. (450°F.).

The following example illustrates the preparation of an adulterated form of the new compound and its unique properties. It is understood, of course, that the material and its method of preparation are exemplary and are not to be considered to limit the invention to the particular molar ratio of ingredients, solids content of the slurry and operating conditions outlined.

#### Example II

A slurry of silica was prepared by mixing finely ground diatomaceous earth with water so that the slurry contained 1.2 pounds of solids per gallon. A slurry of hydrated lime was prepared by mixing hydrated lime with water so that the slurry contained 2.0 pounds of solids per gallon. One hundred twenty-four gallons of diatomaceous earth slurry was pumped into the reactor where it was heated by direct injection steam. The diatomaceous earth slurry was followed by 90 gallons of water to flush the feed lines. Then 46 gallons of lime slurry were pumped into the reactor; this was followed by 15 gallons of flushing water to clear the lines. The reaction vessel was agitated continuously and was held at the desired reaction temperature of 232°C. by direct steam injection heating. The inert gases were vented from the top of the reactor so the pressure in the reactor was at

- 11 -

601158

equilibrium steam pressure increasing to 232°C. or about 410 psig. The slurry was reacted for two hours at 232°C. and then was discharged through a cooler into an appropriate tank at such a rate that the reactor was emptied in about 35 minutes. The solids were filtered from the slurry and were dried and ground. The finished product had the following physical properties:

Bulk density, pcf	6.0
Gardner Coleman adsorption (H <sub>2</sub> O)	425 pounds per 100 pounds solids

It should be appreciated that the feed slurry could be heated by means of heat exchangers and the reactor could be heated by alternate means such as a heating coil, electrical heating, etc. Also, the reactants can be added separately as described or together. The necessary elements of the process simply require bringing together the proper amounts of a reactive silica and hydrated lime in a water medium within a reactor, preferably with agitation, and reacting the same at a temperature of at least 182°C., preferably about 232°C., for the required reaction time.

Due to its unique and novel physical and chemical properties, the new compound in either its pure or adulterate form, among other advantageous applications or uses, is especially suitable for use as a carrier in the preparation of a wettable powder insecticide dispersion. The following example illustrates the preparation of a 75% wettable DDT powder employing the new product CS-55 prepared as described in Example II:

- 12 -

601158

Example III

A 75% wettable DDT powder was prepared by dry blending the following ingredients:

375 grams DDT, Pentech H, ground  
 50 grams Barden Clay, a kaolinite clay  
 60 grams CS-55  
 7.5 grams Polyfon P, sodium lignosulfonate  
 7.5 grams Igepon T-77, sodium-N-methyl-N-octadecyl taurate

The foregoing were ground in a 2 inch Reduction Engineering air jet mill, and the powder was subjected to tropical storage such as described in FOA No. 71554-2 (Modified October 1954) specification for DDT water dispersible powder insecticide concentrates, (FOA Interim Specification for 75% DDT Water Dispersible Powder). The powder from the tropical storage test was dispersed in standard 342 ppm hard water and was subjected to a suspendability test as described in FOA No. 71554-2 (Modified October, 1954) specification. A similar 75% DDT powder was made up and tested using calcium silicate hydrate I prepared in accordance with the teachings of Calvert. The results are as follows:

	% DDT in Suspension After the Tropical Storage Test
CS-55	1.5
Calcium Silicate Hydrate I	0.9

Inasmuch as the USA-FOA specifications require the powder to have a suspendability of 1.2% or more after tropical storage, it is obvious that CS-55 is suitable and that calcium silicate hydrate I is not.

- 13 -

601158

1 The new hydrous calcium silicate product is similarly  
2 suitable for use as a carrier for numerous other powdered  
3 insecticides, such as, for example, Shell Chemical Corpora-  
4 tion's Technical Aldrin containing at least 77.9% 1,2,3,4,  
5 10,10-hexachloro-1,4,5,8,8a-hexahydro-1, 4-endo, exo-5,8-  
6 dimethanonaphthalene (HMDN) and Dieldrin containing at least  
7 85% 1,2,3,4,10,10-hexachloro-6,7, epoxy-1,4,4a,5,6,7,8,8a-  
8 octahydro-1, 4-endo, exo-5,8-dimethanonaphthalene (HMOD);  
9 Aramite containing at least 90% 2-(p-tert-butylphenoxy) iso-  
10 propyl-2-chloroethyl sulfite manufactured by the Naugatuck  
11 Chemical Division of the United States Rubber Company, or  
12 Toxaphene, a chlorinated camphene ( $C_{15}H_{11}Cl_9$ ) manufactured by  
13 Hercules Powder Co.

14 A still further striking difference between the product  
15 produced as described by this invention and that described by  
16 Calvert is the filtration rate of the reacted slurry.  
17 Calvert claims that the product produced according to his  
18 patent is a free filtering product which is a good filter  
19 aid. It has been confirmed that calcium silicate hydrate I  
20 prepared according to the patent has free filtering qualities.  
21 However, the new product which is described in this invention  
22 is not free filtering. Data substantiating this are as  
23 follows:

#### 24 Example IV

25 One-half gallon of the reacted slurry containing about  
26 0.5 pounds solids per gallon was filtered on a Buchner  
27 filter. The slurry temperature was 71 to 77°C. and the  
28 vacuum was 27 inches of mercury. The product, CS-55, pro-  
29 duced as described in Example II required 200 seconds to  
30 filter. A slurry containing calcium silicate hydrate I  
31 required only 35 seconds to filter.  
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- 14 -

601158

1 One of the most important differences between the pro-  
2 duct described by Calvert and that described by this inven-  
3 tion is the difference in solubility. It is believed that  
4 this to a great extent affects the suitability of the product  
5 for use in insecticide formulations and other applications.  
6 Inerts with low solubilities have less tendency to cause  
7 flocculation. The solubility of calcium silicate hydrate I  
8 and CS-55 were determined by mixing an excess of the calcium  
9 silicate solids with distilled water and allowing the mixture  
10 to stand for 4 days. The solids were then filtered from the  
11 slurry, and the filtrate was analyzed. The filtrate which  
12 had been in contact with the calcium silicate hydrate I  
13 contained 101 ppm of dissolved solids. The filtrate which  
14 had been in contact with the new calcium silicate described  
15 by this invention contained only 39 ppm of dissolved solids.  
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- 15 -

601158

SUPPLEMENTARY DISCLOSURE

Whereas the principal disclosure concerns a new hydrated calcium silicate, products thereof, and a method of producing the same, subsequent investigations have produced further data more completely establishing or fixing the scope of the method of preparing the new hydrated calcium silicate of the principal disclosure and products thereof, and encompassing the boundaries of the various reaction conditions comprising said method. Accordingly, this supplementary portion of the disclosure is specifically directed only to the completion of the disclosure insofar as defining substantially all practical and feasible reaction conditions such as periods of reaction and/or temperatures thereof capable of successfully producing the novel hydrated calcium silicate, and products thereof, of the said principal disclosure. However, in that the new hydrated calcium silicate compound, and the products thereof, appear to be capable of precise identification and/or description only through the recitation of certain of their physical properties and/or specific conditions of their hydrothermal conception, the more complete current definition of the method which comprises the basis of this latter disclosure has been also utilized to render more definite and exacting the identification and delineation of the hydrated calcium silicate compound of the principal disclosure and products thereof.

Fig. 4 is a graph defining the time and temperature reaction conditions necessary in the preparation of the new hydrous calcium silicate of the present invention, and illustrates the accelerating effect of agitation upon said reaction conditions.

- 16 -

601158

Fig. 5 is a graph showing the dehydration curve of the hydrous calcium silicate truscottite for comparison with the dehydration characteristics of the novel hydrous calcium silicate compound of this invention.

As set forth above, the preparation of the new hydrated calcium silicate of this invention, and products thereof, may be effected by hydrothermally reacting lime and reactive siliceous material as starting materials in relative proportions of mol ratios of about 0.67 mols of CaO to 1 mol of  $\text{SiO}_2$  for the hydrous calcium silicate compound, or within the approximate range consisting of about 0.05 to 0.7 mols of CaO to 1 mol of  $\text{SiO}_2$  for products comprising the same hydrous calcium silicates. It is essential, of course, that water also be present during the reaction at least in an amount sufficient to provide the chemically combined water indicated in the formula. Moreover, as should be apparent, the amount of water mixed with the starting materials, or present during the reaction, should be sufficient to produce a flowable mass of ambient reactants whereby uninhibited and ready contact of substantially all of said reactants facilitates the reaction.

In addition to the foregoing relative proportions or mol ratios of lime, siliceous material and water as starting materials, specific temperature conditions over definite

- 17 -

601158

1 periods of time are essential to effect the necessary hydro-  
2 thermal reaction of the lime, siliceous material and water  
3 ingredients to form the particular hydrous calcium silicate,  
4 or products thereof, of this invention. Understandably, the  
5 reaction temperature and period or duration thereof, are to  
6 an extent, interdependent conditions, i.e., the higher the  
7 reaction temperature the shorter the period of exposure to  
8 said temperatures and, conversely, the longer the period  
9 of exposure to reaction temperatures the lower the tempera-  
10 ture necessary to effect the hydrothermal formation of the  
11 hydrous silicate compound or products thereof.

12 The slurry or suspension of lime and reactive silica,  
13 prepared in accordance with the foregoing, is then hydro-  
14 thermally reacted for a period of time, and at a temperature  
15 within the approximate limits defined by the solid lines  
16 designated A and B in the accompanying graph of Fig. 4 to  
17 effect the desired reaction. Agitation of the slurry con-  
18 taining the reaction materials, however, substantially  
19 increases the reaction rate over a wide temperature range  
20 and thereby materially reduces the required reaction time  
21 over that range of temperatures and this accelerating effect  
22 of agitation upon the reaction, manifested in a reduction  
23 in time and/or temperature required to effect the reaction  
24 over a certain range of the permissible time and temperature  
25 reaction conditions, is illustrated and approximately defin-  
26 ed by the broken line C and the solid line A in the graph of  
27 Fig. 4. It is accordingly to be understood that the  
28 reaction conditions may comprise any reaction period (time)  
29 and/or temperature within the area(s) defined by the solid  
30 lines A and B, or the solid line B and the broken line C of  
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- 18 -

601158

1 the graph of Fig. 4 and time-temperature reaction conditions  
2 defined within the said area of the graph defined by the  
3 solid line A and the broken line C, when accompanied by  
4 agitation, are therefore considered to be within the scope  
5 of this invention, as well as the time-temperature reaction  
6 conditions within the area defined by the solid lines A and  
7 B. The preferred time and temperature reaction conditions,  
8 either with or without agitation, are illustrated and  
9 defined by the dotted lines D, which set forth the principal  
10 area of most efficient and economical operating conditions.  
11 It will be noted that insufficient reaction periods and/or  
12 temperatures, i.e., reaction conditions within area to the  
13 left of the lines C or A in the graph of Fig. 4 depending  
14 upon whether or not agitation is employed, fail to produce  
15 the new calcium silicate hydrate of the instant invention,  
16 but typically result in calcium silicate hydrate I, a com-  
17 pound of variable composition referred to hereinbefore.  
18 Moreover, it has been found that over reacting, i.e.,  
19 reaction periods and/or temperatures in the area to the  
20 right of or in excess of those defined by the solid lines A  
21 and B in the graph of Fig. 4, can result in calcium sili-  
22 cates of substantially different composition and physical  
23 and chemical properties or characteristics from the new cal-  
24 cium silicate compound, or products thereof, of the present  
25 invention. For example, certain time and/or temperature  
26 reaction conditions in excess of those indicated in the  
27 graph of Fig. 4, i.e., to the right of the necessary reaction  
28 area, have been found to produce, among other materials, com-  
29 pounds which simulate, or comprise the mineral truscottite  
30 and/or the mineral xonotlite, depending, of course, upon the  
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- 19 -

601158

CaO/SiO<sub>2</sub> molar ratio provided by the components, the temperature and/or time of exposure to the same.

Upon X-ray analysis CS-55 manifests the following unique and characteristic pattern as compared to the X-ray diffraction patterns for calcium silicate hydrate I and the minerals gyrolite and truscottite. The gyrolite and truscottite patterns are reproduced from X-ray data found in Heller and Taylor, Crystallographic Data for the Calcium Silicates, Department of Scientific and Industrial Research, published by Her Majesty's Stationary Office, London, 1956.

Angle 2θ	d in Å	CSHI Intensity	CS-55 Intensity	Gyrolite Intensity	Truscottite Intensity
3.7	23.86	-	M	-	-
3.84	22.99	-	-	VS	-
4.6	19.00	-	-	-	S
7.9	11.18	S	M	VS	-
8.2	10.77	-	M	-	-
9.4	9.40	-	W	-	S
10.6	8.34	-	M	-	-
11.2	7.89	-	W	M	-
11.6	7.65	-	-	W	M
14.0	6.30	-	-	W	W
16.1	5.50	S	-	-	-
17.4	5.09	-	-	-	-
17.6	5.02	-	-	-	VW
18.7	4.95	-	-	W	-
19.0	4.65	-	M	M	MW
19.4	4.57	-	-	-	-
19.9	4.46	-	-	-	-
20.3	4.37	-	-	-	-

- 20 -

601158

	Angle 2 $^{\circ}$	d in Å	CSHI Intensity	CS-55 Intensity	Gyrolite Intensity	Truscottite Intensity
1						
2						
3	21.1	4.20	-	-	S	-
4	21.2	4.12	-	VS	-	-
5	21.5	4.13	-	-	-	B
6	22.0	4.04	-	W	-	-
7	22.5	3.95	-	-	-	-
8	23.4	3.80	-	-	-	MS
9	24.0	3.73	-	-	W	W
10	24.4	3.65	-	W	MS	-
11	25.5	3.49	W	W	-	-
12	25.7	3.47	-	-	W	MS
13	26.7	3.34	-	-	-	W
14	27.1	3.29	W	-	-	-
15	28.0	3.18	-	-	K	-
16	28.4	3.14	-	-	-	S
17	28.6	3.12	-	VS	VS	-
18	29.0	3.08	VS	-	-	-
19	29.4	3.04	-	VS	W	-
20	29.8	3.00	-	-	-	K
21	29.9	2.99	-	-	-	-
22	30.0	2.98	VS	-	-	-
23	30.9	2.89	-	-	-	-
24	31.6	2.83	-	-	-	S
25	31.8	2.81	W	W	MS	-
26	32.1	2.79	-	W	-	-
27	32.14	2.78	-	W	M	-
28	33.16	2.70	-	-	M	-
29	34.0	2.64	-	S	-	-
30	34.1	2.63	-	-	-	S
31						
32						
33						

- 21 -

601158

			CSH1	CS-55	Gyrolite	Truscottite
			Intensity	Intensity	Intensity	Intensity
1						
2	<u>Angle 2<math>\theta</math> in <math>^{\circ}</math></u>					
3	35.7	2.51	W	W	-	W
4	36.1	2.48	-	VW	-	VW
5	36.9	2.43	-	M	W	VW
6	37.2	2.42	-	-	W	VW
7	37.3	2.41	W	-	-	-
8	39.5	2.28	M	M	W	-
9	39.9	2.26	VW	-	W	W
10	40.4	2.23	-	M	W	W

Key

VS	-	Very Strong
S	-	Strong
M	-	Medium
W	-	Weak
VW	-	Very Weak

The differential thermal analysis patterns for CS-55 and calcium silicate hydrate I, obtained at a temperature increase of 12.5°C. per minute, are illustrated in Fig. 1 of the drawing. The patterns clearly illustrate the differences in the approach to, and the breaking points of the two compounds and their subsequent formation of two different compounds.

The dehydration curves for CS-55, calcium silicate hydrate I, gyrolite and truscottite are shown in Figs. 2 and 5 of the drawing. The gyrolite and truscottite curves each correspond to the dehydration curves of Fig. 2, page 454 of the A. L. Mackay and H. F. W. Taylor article, "Truscottite", The Mineralogical Magazine and Journal of the Mineralogical Society, Vol. XXX - No. 226, September 1954. The thermal dehydration of CS-55 produces a rapid and substantially complete loss of water at low temperature, i.e., an initial



- 22 -

601158

weight loss of approximately 8% within the first 100°C. temperature increase, an overall loss of approximately 9% at 200°C. and a total weight loss of approximately 10% (10 to 11%) throughout the temperatures ranging from approximately 200° up to 1000°C., as is evidenced by its maintenance of an approximate constant weight throughout said temperature range. The foregoing dehydration characteristic is fully illustrated by the CS-55 curve of Fig. 2. Calcium silicate hydrate I exhibits a dehydration curve comprising a sudden and rapid loss of water at low temperatures, i.e., up to 200°C., a leveling off or plateau indicating little water loss and thus stability between the approximate temperatures of 200 to 400°C. initially not unlike that of CS-55. However, unlike CS-55, the calcium silicate hydrate I curve makes a second sharp rise, indicating a further rapid loss of water, between 400 and 600°C. before again leveling off, after a total water loss of approximately 16% by weight, and maintaining a substantially constant weight from 600 up to 1000°C. The dehydration of gyrolite results in a substantially rapid and uniform initial loss of water which gradually decreases as the temperature increases, with a minor plateau between 300 and 500°C., over the temperature range of 0 to 800°C. before finally leveling off at about 800°C. as indicated. The truscottite dehydration curve exhibits a very gradual and substantially uniform water loss throughout temperatures ranging up to approximately 800°C. when the curve levels off at a water loss represented by a reduction in weight of about 5.5% and continues without further substantial loss up to 1000°C. Thus, the relative stability of CS-55, as

- 23 -

601158

1 evidenced by its rapid initial loss of water at low tempera-  
2 tures, i.e., between approximately 100 to 200°C., and its  
3 subsequent maintenance of a substantially constant weight  
4 or water content over the balance of the temperature range,  
5 in comparison with the varying and irregular dehydration  
6 characteristics of calcium silicate hydrate I, gyrolite and  
7 truscottite, is significant.

8 The carbonation rate of CS-55 is compared with that of  
9 calcium silicate hydrate I in the graph illustrated in Fig.  
10 3 of the drawing. The noticeable difference in the carbona-  
11 tion rate of the two types of products is clearly illustrat-  
12 ed by the graph.

13 The solubility of the new compound or product, CS-55,  
14 is only 39 parts per million, as compared to a solubility  
15 of 101 parts per million for calcium silicate hydrate I.  
16 This particular characteristic is exceptionally noteworthy  
17 since it renders the product especially adaptable to fulfill  
18 certain requirements.

19 Further, the crystalline structure of the CS-55  
20 compound or product is characteristically a crumbled plate  
21 single phase whereas the crystalline configuration of cal-  
22 cium silicate hydrate I consists of a fibrous or bladed  
23 structure, and the crystalline structures of the minerals  
24 gyrolite and truscottite consist of flat plates.

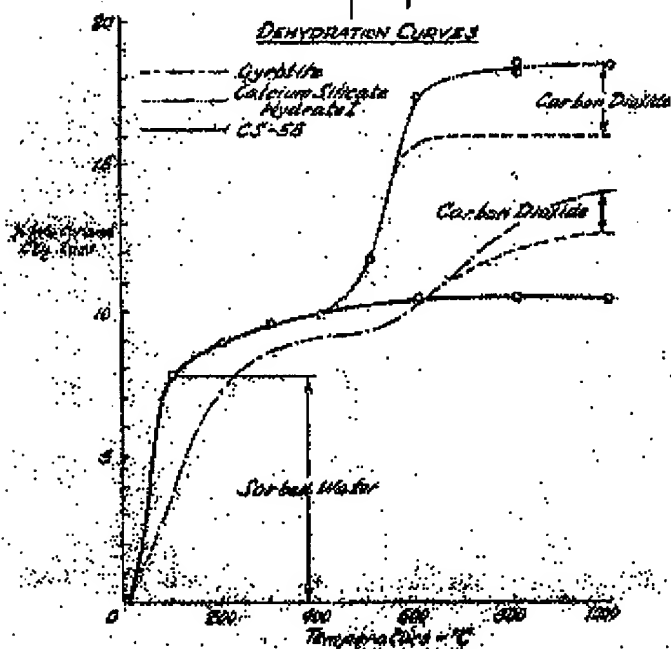
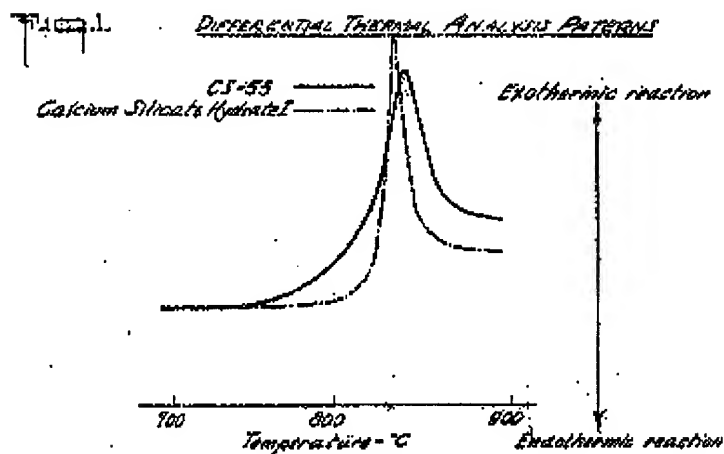
25 Frequently, and in many applications, it is not  
26 necessary to employ the new hydrous calcium silicate CS-55  
27 in its purest form obtainable. Therefore, it is also  
28 within the scope of this invention to prepare and utilize a  
29 product comprising an adulterated form of the compound, or  
30 a mixture of the new compound with hydrated silica and/or  
31  
32  
33

- 24 -

601158

1 possibly other calcium silicate products obtained when lime  
2 and a siliceous material are employed as starting ingredi-  
3 ents in a CaO to SiO<sub>2</sub> molar ratio within the approximate  
4 range of 0.05 to 0.7 CaO to 1 SiO<sub>2</sub> and reacted together as  
5 specified above. In other words, since the advantageous  
6 and unique physical and chemical properties and character-  
7 istics of the product comprising an adulterated composi-  
8 tion, or mixture of the new compound and hydrous silica,  
9 etc., within the foregoing CaO to SiO<sub>2</sub> molar ratio are sub-  
10 stantially similar to those of the pure compound and are  
11 satisfactory for many applications, it is frequently desir-  
12 able and advantageous, particularly for reasons of economy,  
13 to dilute or mix the pure compound with hydrous silica by  
14 including greater quantities of a siliceous material in  
15 the initial slurry than is necessary to obtain pure 2CaO.  
16 3SiO<sub>2</sub>·1-2.5H<sub>2</sub>O.

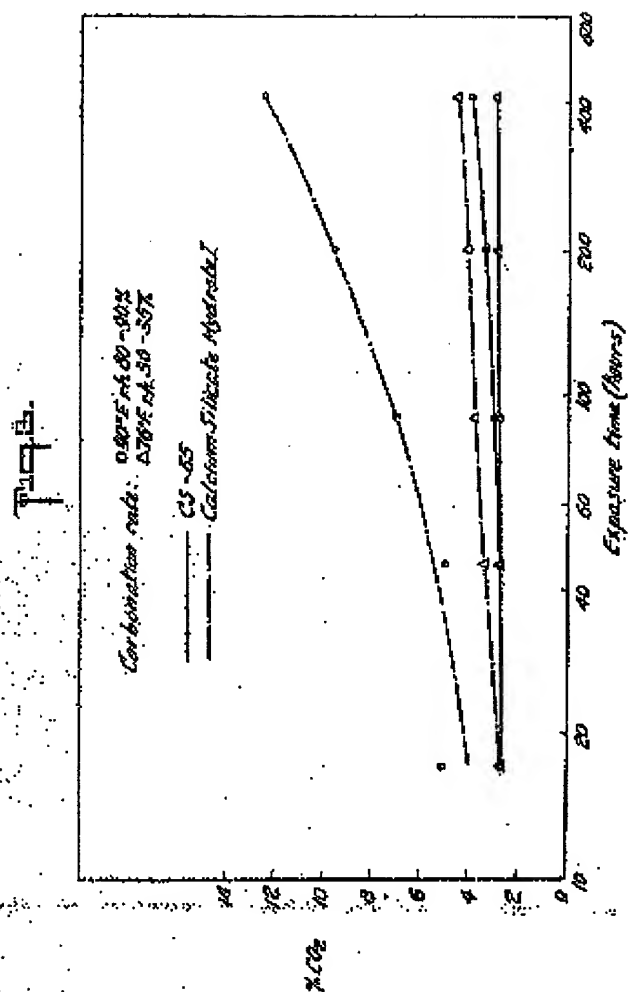
17 Accordingly, it has been found that a satisfactory  
18 product may be prepared by employing a CaO to SiO<sub>2</sub> molar  
19 ratio within the approximate range of 0.05 to 0.7 CaO to  
20 1 SiO<sub>2</sub>, preferably, for commercial purposes, a molar ratio  
21 between 0.4 and 0.6 CaO to 1 SiO<sub>2</sub>. However, in any case,  
22 the reaction temperature and time must be within the  
23 approximate area(s) defined by the solid and/or broken  
24 lines in the graph of Fig. 4.



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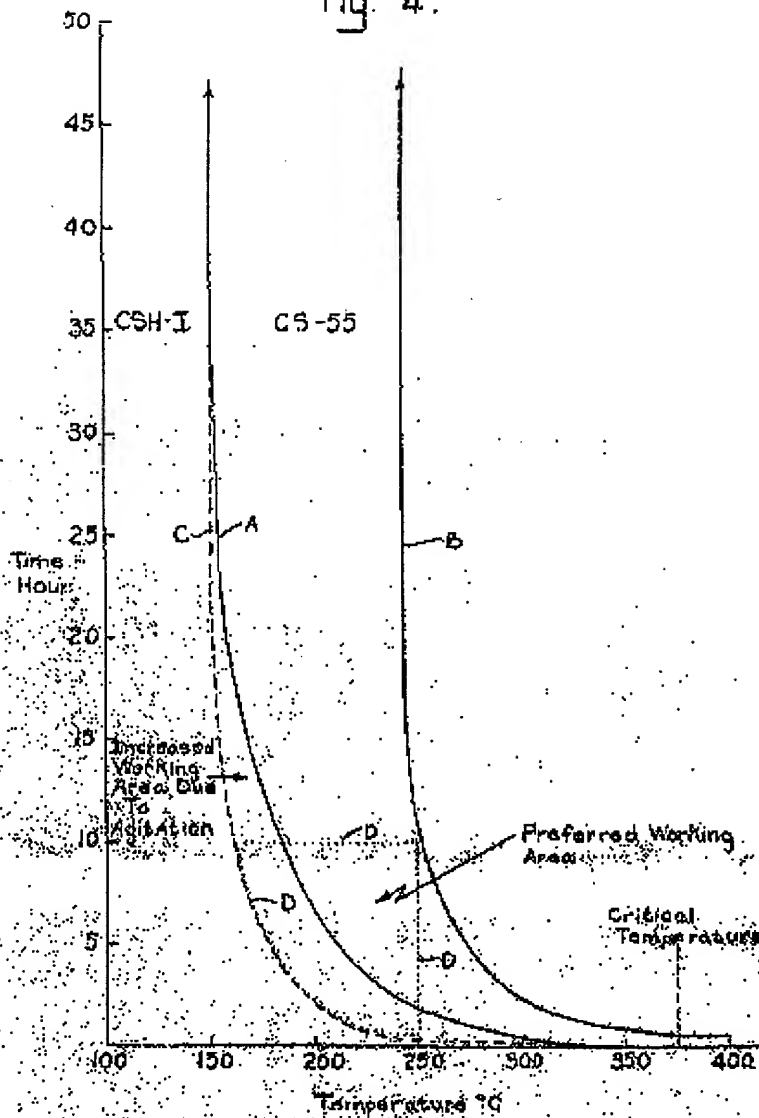


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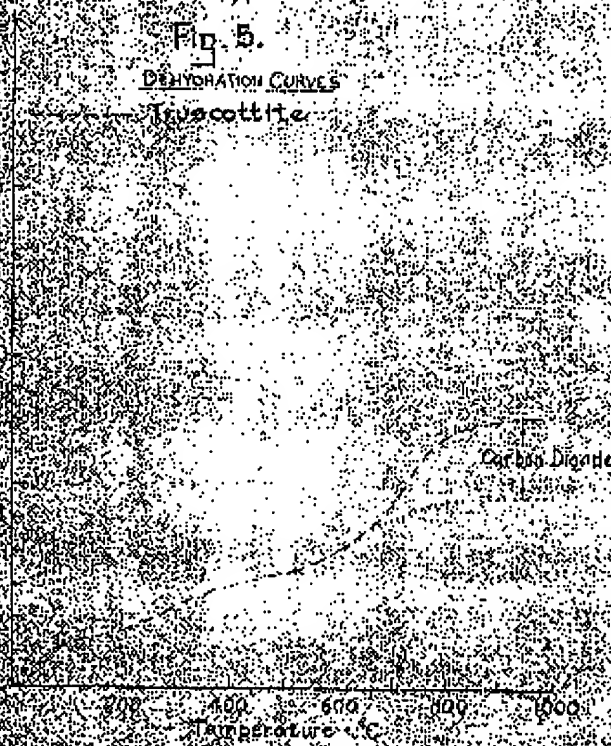
Fig. 4.



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Fig. 5.  
DEHYDRATION CURVES  
Tussockite



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